CLAIMS

1. A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with a labeling reagent according to the Formula

$[Ar_3P^+R]X^-$

wherein

each Ar is an aryl group, all of which may the same or different;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond, thereby linking said analyte to the triarylphosphonium group of the labeling reagent; and

X^- is a negatively-charged counter ion.

2. A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining a triarylphosphonium labeling reagent having a reactive group;

b) obtaining a sample containing an analyte that has an exposed group and that is capable of reacting with said reactive group to thereby form a triarylphosphonium-linked analyte; and

c) reacting said labeling reagent with said analyte such that said triarylphosphonium-linked analyte is formed.

3. The method according to claim 2, wherein said labeling reagent has a structure according to the formula

$[Ar_3P^+R]X^-$

wherein

each Ar is an aryl group, all of which may the same or different;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes; and

X^- is a negatively-charged counter ion.

4. A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with at least two labeling reagents according to the formulae

$$[Ar_3P^+R]X^-$$

and

$$[Ar^*_3P^+R]X^-$$

wherein

Ar and Ar^* are aryl groups, all of which may the same or different, such that the molecular weight of Ar_3P is different from the molecular weight of Ar^*_3P;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming a triarylphosphonium-linked analyte; and

X^- is a negatively-charged counter ion;

such that said triarylphosphonium-linked analyte is formed.

5. A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining at least two triarylphosphonium labeling reagents each having a reactive group, wherein the reactive groups of the labeling reagents are all the same, and the molecular weights of the triarylphosphonium groups of the labeling reagents are different from each other;

b) obtaining a sample containing an analyte that has an exposed group and that is capable of reacting with said reactive group to thereby form a triarylphosphonium-linked analyte; and

c) reacting said labeling reagents with said analyte such that said triarylphosphonium-linked analytes are formed.

6. The method of claim 4, wherein the difference in the molecular weights of the triarylphosphonium groups is discernable by mass spectrometry.

7. The method of claim 4, wherein the difference in the molecular weights of the triarylphosphonium-linked analytes is discernable by mass spectrometry.

8. A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with at least two labeling reagents according to the formulae

$[Ar_3P^+R]X^-$

$[Ar^*_3P^+R]X^-$

$[Ar^{**}_3P^+R]X^-$

. . .

wherein

the Ar groups (*i.e.*, Ar, Ar^*, and Ar^{**}, etc.) are aryl groups, all of which may the same or different, such that the molecular weights of the triarylphosphonium groups of each labeling reagent are unique;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes; and

X^- is a negatively-charged counter ion.

9. A method of analyzing a sample, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group;

b) forming a triarylphosphonium-linked analyte by reacting said analyte with a labeling reagent according to the Formula

$[Ar_3P^+R]X^-$

wherein

each Ar is an aryl group, all of which may the same or different;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming a triarylphosphonium-linked analyte; and

X^- is a negatively-charged counter ion;

such that said triarylphosphonium-linked analyte is formed; and

c) analyzing said triarylphosphonium-linked analyte by mass spectrometry.

10. A method of analyzing a sample, comprising

a) obtaining a triarylphosphonium labeling reagent having a reactive group;

b) obtaining a sample containing an analyte that has an exposed group and that is capable of reacting with said reactive group to thereby form a triarylphosphonium-linked analyte;

c) reacting said labeling reagent with said analyte such that said triarylphosphonium-linked analyte is formed; and

d) analyzing said triarylphosphonium-linked analyte by mass spectrometry.

11. The method according to claim 10, wherein said labeling reagent has a structure according to the formula

$$[Ar_3P^+R]X^-$$

wherein

each Ar is an aryl group, all of which may the same or different;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes; and

X^- is a negatively-charged counter ion.

12. A method of analyzing a sample, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with at least two labeling reagents according to the formulae

$$[Ar_3P^+R]X^-$$

and

$$[Ar^*_3P^+R]X^-$$

wherein

Ar and Ar^* are aryl groups, all of which may the same or different, such that the molecular weight of Ar_3P is different from the molecular weight of Ar^*_3P;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming a triarylphosphonium-linked analyte; and

X^- is a negatively-charged counter ion; and

c) analyzing said triarylphosphonium-linked analyte by a mass spectrometry technique.

13. A method of analyzing a sample, comprising

a) obtaining at least two triarylphosphonium labeling reagents each having a reactive group, wherein the reactive groups of the labeling reagents are all the same, and the molecular weights of the triarylphosphonium groups of the labeling reagents are different from each other;

b) obtaining a sample containing an analyte that has an exposed group and that is capable of reacting with said reactive group to thereby form a triarylphosphonium-linked analyte;

c) reacting said labeling reagents with said analyte such that said triarylphosphonium-linked analytes are formed; and

d) analyzing said triarylphosphonium-linked analyte by a mass spectrometry technique.

14. The method according to claim 13, wherein each of said labeling reagent has a structure according to the formula

$$[Ar_3P^+R]X^-$$

wherein

each Ar is an aryl group, all of which may the same or different, such that the molecular weights of the triarylphosphonium groups of each labeling reagent are unique;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes; and

X^- is a negatively-charged counter ion.

15. The method according to any of the foregoing claims, wherein said mass spectrometry technique is matrix-assisted laser desorption/ionization mass spectrometry or electrospray mass spectrometry.

16. The method according to claim 15, wherein said technique is quantitative.

17. A method of analyzing a sample, comprising

a) obtaining at least two triarylphosphonium labeling reagents each having a reactive group, wherein the reactive groups of the labeling reagents are all the same, and the molecular weights of the triarylphosphonium groups of the labeling reagents are different from each other;

b) obtaining a sample containing an analyte that has an exposed group and that is capable of reacting with said reactive group to thereby form a triarylphosphonium-linked analyte;

c) reacting, in a first vessel, the first labeling reagent with a first portion of said sample such that triarylphosphonium-linked analytes thereof are formed;

d) reacting, in a second vessel, the second labeling reagent with a second portion of said sample such that triarylphosphonium-linked analytes thereof are formed;

e) combining triarylphosphonium-linked analytes from said first vessel with triarylphosphonium-linked analytes from said second vessel to form a mixture; and

f) analyzing said mixture of triarylphosphonium-linked analytes by a mass spectrometry technique.

18. The method of claim 17, further comprising quantitatively comparing the relative signals of the triarylphosphonium-linked analytes from said first vessel to the triarylphosphonium-linked analytes of said second vessel.

19. The method of claim 18, wherein said quantitative comparison is correlated with the relative amounts of the triarylphosphonium-linked analytes.

20. The method of claims 17, wherein the ratio of said first portion of said sample to said second portion of said sample is predetermined and definite.

21. The method of claim 20, wherein said ratio is correlated to the MS signals of the triarylphosphonium-linked analytes.

22. The method of claim 17, wherein the difference in the signals of an analyte labeled with said first labeling reagent and the same analyte labeled with said second labeling reagent is linearly related to the difference in their concentration in said mixture.

23. The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted aryl groups.

24. The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted heteroaryl groups.

25. The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted aromatic hydrocarbons.

26. The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted phenyl, 1-naphthyl, 2-naphthyl, biphenyl, 1-pyrrolyl, 2-pyrrolyl, 3-pyrrolyl, 3-pyrazolyl, 2-imidazolyl, 4-imidazolyl, pyrazinyl, 2-oxazolyl, 4-oxazolyl, 5-oxazolyl, 3-isoxazolyl, 4-isoxazolyl, 5-isoxazolyl, 2-thiazolyl, 4-thiazolyl, 5-thiazolyl, 2-furyl, 3-furyl, 2-thienyl, 3-thienyl, 2-pyridyl, 3-pyridyl, 4-pyridyl, 2-pyrimidyl, 4-pyrimidyl, 5-benzothiazolyl, purinyl, 2-benzimidazolyl, 5-indolyl, 1-isoquinolyl, 5-isoquinolyl, 2-quinoxalinyl, 5-quinoxalinyl, 3-quinolyl, and 6-quinolyl groups.

27. The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted 5– and 6–membered single–ring groups.

28. The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted phenyl, pyrrolyl, furyl, thienyl, thiazolyl, isothiaozolyl, imidazolyl, triazolyl, tetrazolyl, pyrazolyl, oxazolyl, isooxazolyl, pyridinyl, pyrazinyl, pyridazinyl, and pyrimidinyl groups.

29. The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted multicyclic aryl groups.

30. The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted naphthyl, tetrahydronaphthyl, benzoxazolyl, benzodioxazolyl, benzothiazolyl, benzoimidazolyl, benzothiophenyl, methylenedioxyphenyl, quinolinyl, isoquinolinyl, napthyridinyl, indolyl, benzofuranyl, purinyl, deazapurinyl, and indolizinyl groups.

31. The method according to claim 1, wherein two Ar groups together form one divalent aromatic group.

32. The method according to claim 31, wherein divalent aromatic group is selected from the group consisting of substituted or unsubstituted 1,1'-binaphth-2,2'-diyl, phenylene, and xylylene groups.

33. The method according to claim 1, wherein each Ar group is selected from the group consisting of unsubstituted phenyl, unsubstituted naphthyl, unsubstituted indenyl, unsubstituted anthracenyl, substituted phenyl, substituted naphthyl, substituted indenyl, and substituted anthracenyl groups.

34. The method according to claim 33, wherein the substituents are alkyl or alkoxy groups.

35. The method according to claim 33, wherein the substituents are selected from the group consisting of halogens; C_1-C_6 alkyl groups; (C_1-C_4 alkoxy)-substituted C_1-C_6 alkyl groups; C_1-C_6 alkoxy groups; C_1-C_6 alkylthio groups; C_1-C_6 alkanoyl groups; C_1-C_6 alkanoyloxy groups; and C_1-C_6 alkoxycarbonyl groups.

36. The method according to claim 33, wherein the substituents are C_1-C_4 alkyl groups or C_1-C_4 alkylalkoxy groups.

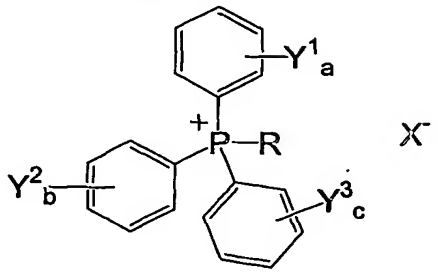
37. The method according to claim 1, wherein said Ar_3P group is selected from the group consisting of substituted or unsubstituted triphenylphosphine, naphthyldiphenylphosphine, dinaphthylphenylphosphine, trinaphthylphosphine, 9-anthryldiphenylphosphine, 9-anthryldinaphthylphosphine, diphenylpyrenylphosphine, dinaphthylpyrenylphosphine.

38. The method according to claim 37, wherein said naphthyl is 1-naphthyl or 2-naphthyl.

39. The method according to claim 37, wherein the substituents are selected from the group consisting of halogens; C_1-C_6 alkyl groups; (C_1-C_4 alkoxy)-substituted C_1-C_6 alkyl groups; C_1-C_6 alkoxy groups; C_1-C_6 alkylthio groups; C_1-C_6 alkanoyl groups; C_1-C_6 alkanoyloxy groups; and C_1-C_6 alkoxycarbonyl groups.

40. The method according to claim 37, wherein said substituted triphenylphosphine is tri(*p*-methoxyphenyl)phosphine.

41. The method according to claim 1, wherein said labeling reagent has a structure according to the formula



wherein

P is a phosphorous atom;

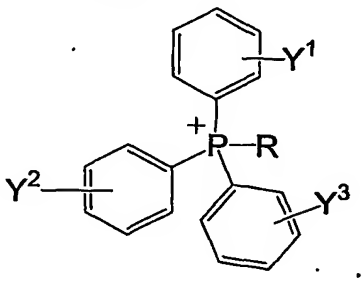
R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes;

a, b, and c are independently integers from 0 to 5;

Y^1, Y^2, and Y^3, which may be the same or different, are independently selected from the group consisting of halogen, hydroxyl, alkylcarbonyloxy, arylcarbonyloxy, alkoxycarbonyloxy, aryloxycarbonyloxy, carboxylate, alkylcarbonyl, alkoxycarbonyl, aminocarbonyl, alkylthiocarbonyl, alkoxyl, phosphate, phosphonato, phosphinato, cyano, amino (including alkyl amino, dialkylamino, arylamino, diarylamino, and alkylarylamino), acylamino (including alkylcarbonylamino, arylcarbonylamino, carbamoyl and ureido), amidino, imino, sulfhydryl, alkylthio, arylthio, thiocarboxylate, sulfate, sulfonato, sulfamoyl, sulfonamido, nitro, trifluoromethyl, cyano, azido, heterocyclyl, aralkyl, aryl, and heteroyl groups, provided that none of said Y groups reacts with said R group; and

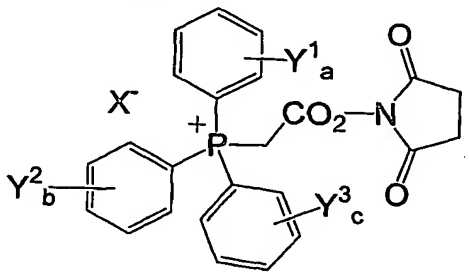
X^- is a negatively-charged counter ion.

42. The method according to claim 1, wherein said labeling reagent has a structure according to the formula

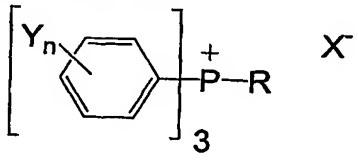


43. The method according to claim 42, wherein Y^1, Y^2, and Y^3 are selected from the group consisting of methyl, ethyl, propyl, isopropyl, butyl, isobutyl, pentyl, hexyl, heptyl, methyloxy, ethyloxy, propyloxy, isopropyloxy, butyloxy, isobutyloxy, pentyloxy, hexyloxy, and heptyloxy.

44. The method according to claim 1, wherein said labeling reagent has a structure according to the formula



45. The method according to claim 1, wherein said labeling reagent has a structure according to the formula



wherein

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes;

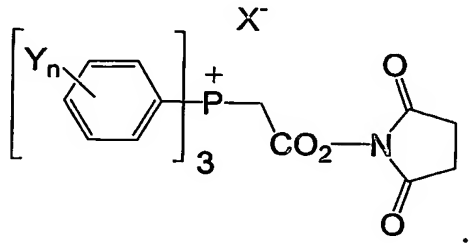
n is an integer from 0 to 5;

Y is selected from the group consisting of halogen, hydroxyl, alkylcarbonyloxy, arylcarbonyloxy, alkoxycarbonyloxy, aryloxycarbonyloxy, carboxylate, alkylcarbonyl, alkoxycarbonyl, aminocarbonyl, alkylthiocarbonyl, alkoxyl, phosphate, phosphonato, phosphinato, cyano, amino (including alkyl amino, dialkylamino, arylamino, diarylamino, and alkylarylamino), acylamino (including alkylcarbonylamino, arylcarbonylamino, carbamoyl and ureido), amidino, imino, sulfhydryl, alkylthio, arylthio, thiocarboxylate, sulfate, sulfonato, sulfamoyl, sulfonamido, nitro, trifluoromethyl, cyano, azido, heterocyclyl, aralkyl, aryl, and heteroyl groups, provided that none of said Y groups reacts with said R group; and

X^- is a negatively-charged counter ion.

46. The method according to claim 44, wherein Y^1, Y^2, and Y^3 are selected from the group consisting of methyl, ethyl, propyl, isopropyl, butyl, isobutyl, pentyl, hexyl, heptyl, methyloxy, ethyloxy, propyloxy, isopropyloxy, butyloxy, isobutyloxy, pentyloxy, hexyloxy, and heptyloxy.

47. The method according to claim 1, wherein said labeling reagent has a structure according to the formula



48. The method according to claim 2, wherein each of said triarylphosphonium labeling reagent has the same chemical structure, and wherein each triarylphosphonium labeling reagent is isotopically enriched with respect to the other triarylphosphonium labeling reagents.

49. The method according to claim 48, wherein a triarylphosphonium labeling reagent isotopically enriched with ^{12}C, ^{12}C, ^{1}H, or ^{2}H.

50. The method according to claim 41, wherein Y^1, Y^2, and Y^3 are selected from the group consisting of $O^{12}C^{1}H_3$, $O^{12}C^{2}H_3$, $O^{13}C^{1}H_3$, and $O^{13}C^{2}H_3$.

51. The method according to claim 1, wherein said exposed group of said analyte is electrophilic and said reactive functional group is nucleophilic.

52. The method according to claim 1, wherein said exposed group of said analyte is nucleophilic and said reactive functional group is electrophilic.

53. The method according to claim 1, wherein said exposed group of said analyte is an amine, and said reactive group comprises an isothiocyanate, succinimidyl esters, or aldehyde.

54. The method according to claim 1, wherein said exposed group of said analyte is an alcohol or thiol, and said reactive group comprises an alkylating group.

55. The method according to claim 54, wherein said alkylating group is an iodoacetamide, maleimide, or alkyl halide.

56. The method according to claim 1, wherein X^- is a halide, triflate, sulfate, nitrate, hydroxide, carbonate, bicarbonate, acetate, phosphate, oxalate, cyanide, aklylcarboxylate, *N*-hydroxysuccinimide , *N*-hydroxybenzotriazole, alkoxide, thioalkoxide, alkane sulfonyloxy, halogenated alkane sulfonyloxy, arylsulfonyloxy, bisulfate, oxalate, valerate, oleate, palmitate, stearate, laurate, borate, benzoate, lactate, citrate, maleate, fumarate, succinate, tartrate, naphthylate mesylate, glucoheptonate, or lactobionate.

57. The method according to claim 1, wherein X^- is an anionic Y group such that the labeling reagent is zwitterionic.

58. A composition comprising at least two different labeling reagents each having a different molecular weight according to the formula

$$[Ar_3P^+R]X^-$$

wherein

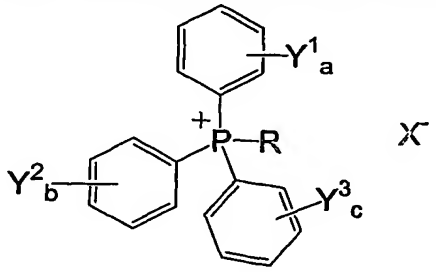
each Ar is aryl group, all of which may the same or different;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming a triarylphosphonium-linked analyte; and

X^- is a negatively-charged counter ion.

59. A composition comprising at least two different labeling reagents each having a different molecular weight according to the formula



wherein

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes;

a, b, and c are independently integers from 0 to 5;

Y^1, Y^2, and Y^3, which may be the same or different, are independently selected from the group consisting of halogen, hydroxyl, alkylcarbonyloxy, arylcarbonyloxy, alkoxycarbonyloxy, aryloxycarbonyloxy, carboxylate, alkylcarbonyl, alkoxycarbonyl, aminocarbonyl, alkylthiocarbonyl, alkoxyl, phosphate, phosphonato, phosphinato, cyano, amino (including alkyl amino, dialkylamino, arylamino, diarylamino, and alkylarylamino), acylamino (including alkylcarbonylamino, arylcarbonylamino, carbamoyl and ureido), amidino, imino, sulfhydryl, alkylthio, arylthio, thiocarboxylate, sulfate, sulfonato, sulfamoyl, sulfonamido, nitro, trifluoromethyl, cyano, azido, heterocyclyl, aralkyl, aryl, and heteroyl groups, provided that none of said Y groups reacts with said R group; and

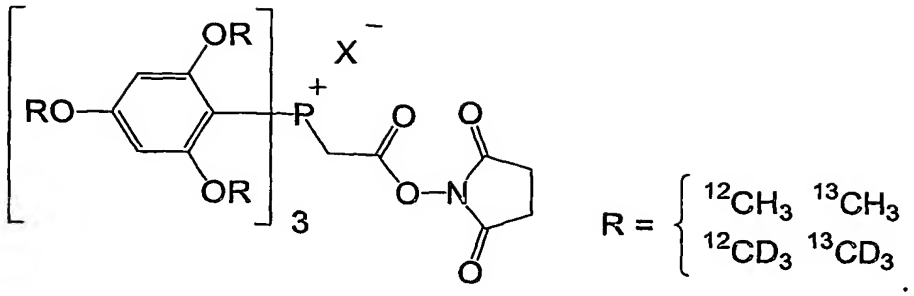
X^- is a negatively-charged counter ion.

60. The composition according to claim 59, wherein each labeling reagent has the same chemical structure, and wherein each labeling reagent is isotopically enriched with respect to the other labeling reagents.

61. The method according to claim 60, wherein a labeling reagent isotopically enriched with ^{12}C, ^{13}C, ^{1}H, or ^{2}H.

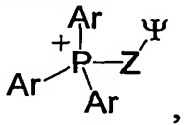
62. The method according to claim 59, wherein Y^1, Y^2, and Y^3 are selected from the group consisting of $O^{12}C^1H_3$, $O^{12}C^2H_3$, $O^{13}C^1H_3$, and $O^{13}C^2H_3$.

63. The method according to claim 1, wherein the labeling reagent has the following structure:



64. The method according to claim 1, wherein the labeling reagent has the following structure:

65. The method according to claim 1, wherein the labeling reagent has the following structure:



wherein

each Ar is aryl group, all of which may the same or different;

P is a phosphorous atom;

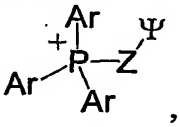
Z is a linking group; and

Ψ is a reactive functional group.

66. The method according to claim 65, wherein said reactive functional group is an activated ester of the formula –COL, where L is a leaving group.

67. The method according to claim 66, wherein L is a *N*-hydroxysulfosuccinimidyl, *N*-hydroxysuccinimidyl, or substituted aryloxy group.

68. The method according to claim 1, wherein the labeling reagent has the following structure:



wherein

each Ar is aryl group, all of which may the same or different;

P is a phosphorous atom;

Z is a linking group; and

Ψ is a reactive functional group.

69. The method according to claim 68, wherein said aryl groups are unsubstituted or substituted with substituents selected from the group consisting of halogens, trifluoromethyl, nitro, cyano, C_1-C_6 alkyl, C_2-C_6 alkenyl, C_2-C_6 alkynyl, C_1-C_6 alkylcarbonyloxy, arylcarbonyloxy, C_1-C_6 alkoxycarbonyloxy, aryloxycarbonyloxy, C_1-C_6 alkylcarbonyl, C_1-C_6 alkoxycarbonyl, C_1-C_6 alkoxy, C_1-C_6 alkylthio, arylthio, heterocyclyl, aralkyl, and aromatic and heteroaromatic groups.

70. The method according to claim 65, wherein said Ψ group is a carboxylic acid, a derivative of a carboxylic acid, or an activated ester of a carboxylic acid.

71. The method according to claim 65, wherein said Ψ group is a haloalkyl, haloacetamide, halomethylbenzamide, a maleimido group, or a sulfonate ester, wherein the sulfonic acid is an alkylsulfonic acid, perfluoroalkylsulfonic acid, or an arylsulfonic acid.

72. The method according to claim 65, wherein said Ψ group is an iodoacetamide, maleimide, or a halomethylbenzamide.

73. The method according to claim 65, wherein said Ψ group is an isocyanate or an acyl nitrile.

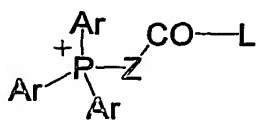
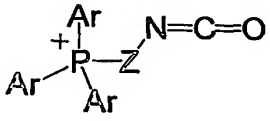
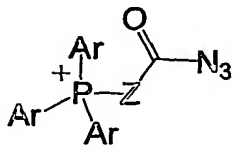
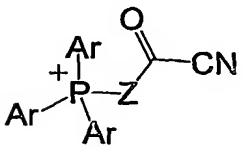
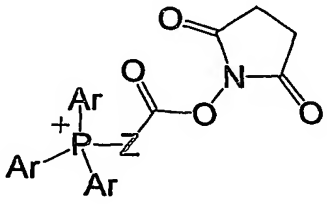
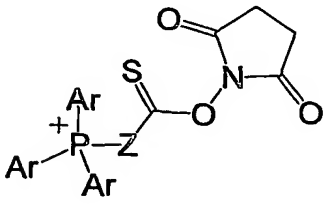
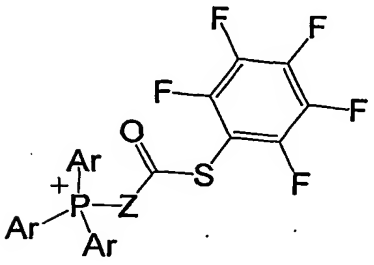
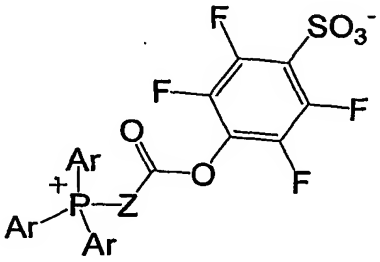
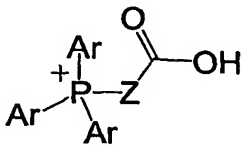
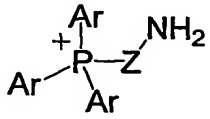
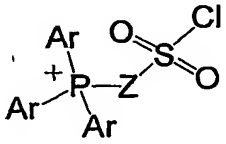
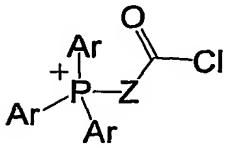
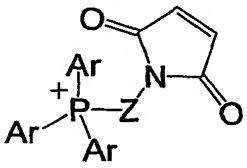
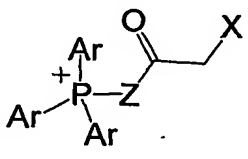
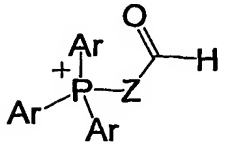
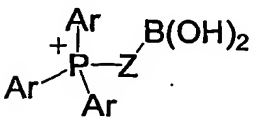
74. The method according to claim 65, wherein said Ψ group is a carboxylic acid, an activated ester of a carboxylic acid, an acyl azide, an acyl halide, a symmetric or asymmetric anhydride, an acrylamide, an alcohol, a thiol, an aldehyde, an amine, an azide, an imido ester, a sulfonate ester, a haloacetamide, an alkyl halide, a sulfonyl halide, a hydrazine, an isocyanate, an isothiocyanate, or a maleimide group.

75. The method according to claim 65, wherein said Ψ group is a carboxylic acid, a succinimidyl ester, an amine, a haloacetamide, an alkyl halide, a sulfonyl halide, an isothiocyanate, or a maleimide group.

76. The method according to claim 65, wherein said Ψ group is a succinimidyl ester.

77. The method according to claim 65, wherein said Ψ group is an acyl azide, an acyl nitrile, an aldehyde, an alkyl halide, an amine, an anhydride, an aniline, an aryl halide, an azide, an aziridine, a boronate, a carboxylic acid, a diazoalkane, a haloacetamide, a hydrazine, an imido ester, an isocyanate, an isothiocyanate, a maleimide, a sulfonyl halide, or a thiol group.

78. The method according to claim 65, wherein said labeling reagent has a structure selected from the group consisting of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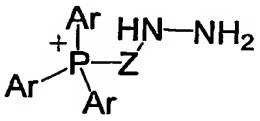


wherein

each Ar is aryl group, all of which may the same or different;

P is a phosphorous atom;

Z is a linking group; and

X is a leaving group; and

L is a leaving group.

79. The method according to claim 1, wherein Z has 1-20 nonhydrogen atoms selected from the group consisting of C, N, O and S, and the longest linear segment contains 1-6 nonhydrogen atoms.

80. The method according to claim 79, wherein Z is a single methylene group.

81. The method according to claim 79, wherein Z is a polymethylene of the formula $-(CR'R''_2)_n-$, where n is 1 to 10, and where each of R' and R'' are each independently hydrogen, a C_1-C_5 alkyl, C_2-C_5 alkenyl, C_2-C_5 alkynyl, or aryl group.

82. The method according to claim 79, wherein Z is a polymethylene of the formula $-(CH_2)(CR'R''_2)_n-$, where n is 0 to 9, and R' and R'' are each independently hydrogen, a C_1-C_5 alkyl, C_2-C_5 alkenyl, C_2-C_5 alkynyl, or aryl group.

83. The method of claim 1, wherein said analyte is a protein, peptide, enzyme, immunoglobulin, hapten, antigen, amino acid, hormone, receptor, nucleic acid, hormone, chemical, polymer, pathogen, toxin, saccharide or polysaccharide, steroid, vitamin, therapeutic drug, drug of abuse, bacterium or virus, or a combination or fragment of any of the foregoing, or a metabolite thereof, or an antibody thereto.

84. The method of claim 1, wherein said analyte is a food additive, agrichemical, surfactants, adhesives, resin, organic pollutant, or process chemical.

85. The method of claim 1, wherein said analyte is a therapeutic drug or a metabolite thereof.

86. The method of claim 1, wherein said analyte is a drug of abuse or a metabolite thereof.

87. The method of claim 1, wherein said sample is rainwater, or water from an ocean, river, lake, pond, or stream.

88. The method of claim 1, wherein said sample is a biological tissue.

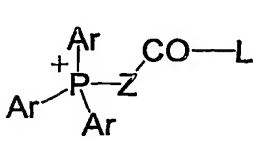
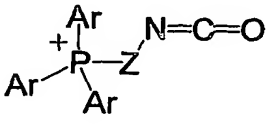
89. The method of claim 1, wherein said sample is whole blood, plasma, serum, urine, cerebrospinal fluid, ascites fluid, sweat, lymph, or other body fluids.

90. A kit for use in preparing a sample for mass spectrometry analysis comprising a labeling reagent according to claim 1, and instructions for use in the method of the instant invention.

91. A kit for use in preparing a sample for mass spectrometry analysis comprising a labeling reagent according to claim 1, and buffer chemicals.

92. A kit according to claim 1, further comprising acids, bases, buffering agents, inorganic salts, solvents, antioxidants, preservatives, or metal chelators, or aqueous or organic solutions thereof.

93. A labeling reagent having a structure selected from the group consisting of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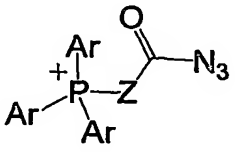
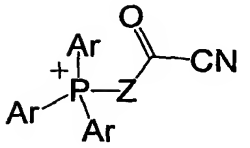
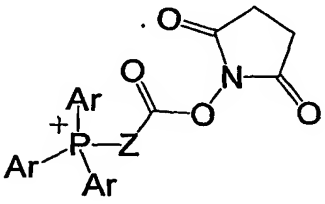


wherein

each Ar is aryl group, all of which may the same or different;
P is a phosphorous atom;
Z is a linking group; and
L is a leaving group.

94. A labeling reagent having a structure selected from the group consisting of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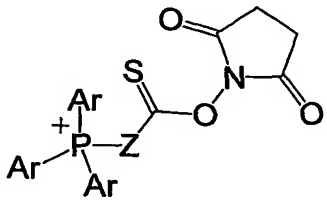
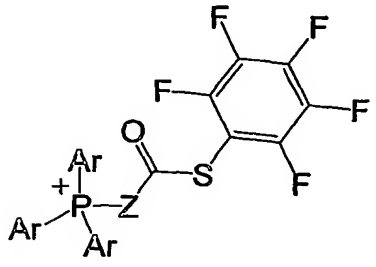
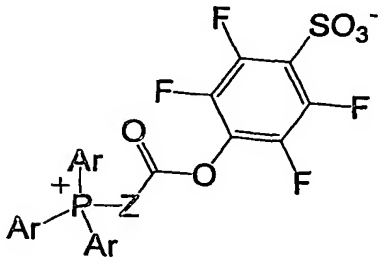


wherein

each Ar is aryl group, all of which may the same or different;
P is a phosphorous atom; and
Z is a linking group.

95. A labeling reagent having a structure selected from the group consisting of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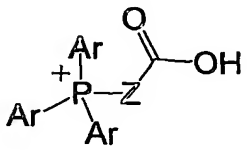


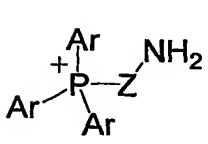
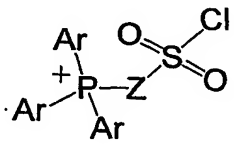
wherein

each Ar is aryl group, all of which may the same or different;

P is a phosphorous atom; and

Z is a linking group.

96. A labeling reagent having a structure selected from the group consisting of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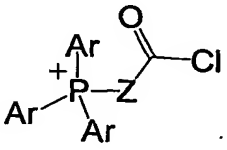


wherein

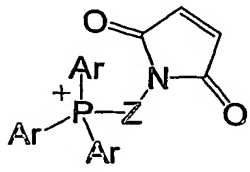
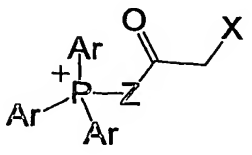
each Ar is aryl group, all of which may the same or different;

P is a phosphorous atom; and

Z is a linking group.

L is a leaving group.

97. A labeling reagent having a structure selected from the group consisting of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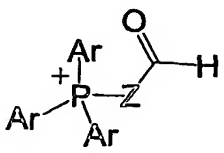

wherein

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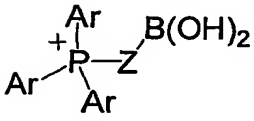
P is a phosphorous atom;
Z is a linking group; and
X is a leaving group.

98. A labeling reagent having a structure selected from the group consisting of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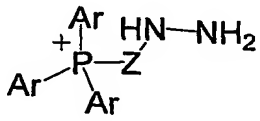

wherein

each Ar is aryl group, all of which may the same or different;
P is a phosphorous atom; and
Z is a linking group.